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Advances in targetry with thin diamond-like carbon foils

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Abstract

Thin and stable diamond-like carbon (DLC) foils, which were fabricated at the Kurchatov Institute by sputter deposition, have proved recently to be advantageous for stripping and secondary electron timing of high energy heavy ions in a number of accelerator experiments. This resulted in expanding applications of these DLC foils which necessitated further development efforts directed toward the following applications of DLC targetry: (i) thin stripper foils for lower energy tandem accelerators, (ii) enlarged (up to 66 mm in diameter) stop foils for improved time-of-flight elastic recoil detection ion beam analysis, and (iii) ultra-thin (about 0.6 μg/cm^2) DLC foils for some fundamental and applied physics experiments. Along with the fabrication of thin DLC stripper foils for tandem accelerators, much thicker (up to 200 μg/cm^2) foils for post-stripping of heavy-ion beams in higher energy linacs, are within reach. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

Thin and stable carbon foils are needed in many particle accelerator experiments on nuclear and atomic physics as well as in some interdisciplinary research. Although a variety of conventional, mostly thermally evaporated, carbon foils are presently commercially available, their irradiation hardness under very heavy-ion irradiation and minimal thickness can be a serious operational limitation.

Self-supporting diamond-like carbon (DLC) foils, which were developed at the Kurchatov Institute [1], have shown lately to be promising as long-lived stripper foils when exposed to high intensity 15 MeV Au ion beams at the terminal of a tandem accelerator (Brookhaven National Lab.) [2]. During more systematic lifetime measurements, performed at the MP tandem of the Max–Planck-Institut für Kernphysik, Heidelberg, a large number of 5 μg/cm^2 DLC foils have been irradiated in comparison with other stripper foils of the same thickness [3] with about 1 μA, 11 MeV Au and Cu ions in a vacuum of $\approx 10^{-5}$ Pa. The latter foils were produced by cracking of ethylene

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[4] and laser plasma ablation [5]. In these experiments, DLC foils exhibited lifetimes longer by a factor of about ten than those of arc-evaporated carbon reference foils, and compared favorably with other advanced foils, tested. Approximately the same improvement factor was obtained in a lower energy range with $3\,\mu g/cm^2$ DLC stripper foils when bombarded with 2.3 MeV ^{58}Ni beams at the Pelletron Accelerator of the University of Lund [6]. High mechanical strength of DLC coupled with low electron affinity made it possible to develop ultra-thin ($0.6\,\mu g/cm^2$) secondary electron-emitting (SE) foils for time-of-flight (ToF) spectrometers. Due to a reduced energy straggling in the start foil the energy resolution of several back scattering ToF spectrometers [7,8] could be significantly improved for high and medium energy ion beam analysis (IBA).

In this article, we report on further development and evaluation efforts motivated by expanding applications of DLC foils in a variety of accelerator experiments. The main features of DLC foil preparation and characterization are presented in detail. It was demonstrated that the DLC stripper foils show excellent quality for very heavy ions with energies above 10 MeV. In this paper, results are described for lifetime tests obtained with 2.3 MeV ^{12}C and 1.5 MeV ^{2}D ion beams. With growing experience in the fabrication of thin DLC stripper foils for tandem accelerators, the preparation of much thicker DLC foils seems to be within reach. They are required for post-stripping of high energy heavy-ion beams, e.g. in high energy linacs.

Recent application of DLC targetry to Elastic Recoil Detection Analysis (ERDA) techniques with improved sensitivity resulted in development DLC stop foils with a large active diameter of 66 mm. The break through for the preparation of such thin and large area DLC foils came with a novel heat treatment technique implemented to mount very flat and mechanically stable supporting mesh.

In the last part of the article, advantages are described when using ultra-thin DLC foils as well for Coulomb explosion imaging (CEI) technique as for the investigation of pre-equilibrium interaction of highly charged ions with solids.

2. Experimental

2.1. Preparation and characterization of DLC foils

The DLC foils have been produced by the special-purpose DC glow discharge sputtering of graphite in a low density krypton plasma. Carbon atoms are condensed onto glass slides which are coated with a release agent and cooled to liquid nitrogen temperature during the deposition. A detailed description as well of the deposition process as of the DLC foil preparation setup is given in Ref. [1,2]. One of its important features is good starting vacuum ($\sim 10^{-6}$ Pa) minimizing effects of adhering surface layers of especially water vapor onto the film during deposition. Such layers were made responsible to increase the effective thickness of conventional very thin carbon foils [9,10]. Characteristic properties of DLC deposits are their extreme hardness, high electrical resistivity and high chemical inertness, making them attractive for protective coatings of metals, which was the initial application of this technique. Diamond-like properties of the produced films have been tested on thick ($100\,\mu g/cm^2$) deposits. Their hardness (~ 20 GP) and low electrical conductivity ($\sim 5 \times 10^{-4}\,\Omega/cm^2$) are within the scatter of published data for a large variety of non-hydrogenated DLC films with sp^3 bonds content of $\approx 40\%$ [11]. After deposition, the DLC foils were floated off in distilled water and mounted on normal frames or on ones with meshed apertures. DLC foils with a nominal thickness of as small as $0.6\,\mu g/cm^2$ can be produced by this technique. The thickness of the films on slides was measured by transmission of light by means of a small optical monochromator, pre-calibrated by weighing of sets of foil samples $> 2\,\mu g/cm^2$. The optical transmission of the foils was measured in the wavelength range of 200–250 nm enabling a high sensitivity for relative thickness measurements of very thin carbon foils [12]. The density of DLC foils was determined by the measurement of their height using a profilometer (Alpha-Step 200, Textor Instruments) and their thickness in units of $\mu g/cm^2$ by weighing with a microbalance. Thus determined density of $20\,\mu g/cm^2$ DLC foils ranges from 2.0 to $2.8\,g/cm^3$

depending on foil deposition conditions. Such measurements of the foil thickness and density have an accuracy of $\pm 20\%$ on average.

Several release agents have been chosen for the preparation of DLC foils. Betaine, protected with an ultra-thin evaporated carbon film [3] was used to produce stripper foils in a thickness range of 1–5 μg/cm^2, strong and flexible enough to withstand subsequent slackening. For the fabrication of thicker DLC foils, which usually require longer deposition times, NaCl (≈ 100 μg/cm^2) has been found more suitable. Such rather thick NaCl release agent films result in a highly corrugated morphology of the foil due to distinct grains of the polycrystalline substrate, replicated by the carbon foil. Such topography of stripper foils was reported to enhance their irradiation hardness [13], which probably is due to additional shrinkable area. Unlike stripping in tandem accelerators, ToF and CEI applications clearly require target foils to be as thin as possible with a minimal non-uniformity even on a microscopic scale [8]. To meet these requirements, a detergent (potassium oleate) was chosen as a release agent. It was applied as an alcohol solution to specifically selected glasses and polished until no visible traces remained. Atomic force microscope (AFM) images of the thinnest mesh supported DLC foils were reported [8]. Although measured rms roughness of detergent-treated slides is <3 nm, AMS image shows significantly larger non-uniformity on a microscopic scale. Assuming that the foil deposition is a random process, one could expect a thickness variation of $\pm 30\%$ for 0.6 μg/cm^2 (≈ 10 atomic layers) foils [14]. However, this essential feature of ultra-thin foils should be investigated more.

Along with the fabrication of thin and very thin DLC foils, we made attempts to extend the thickness range up to 200 μg/cm^2. Thick DLC foils are required for post-stripping of heavy ions in the energy range of 1–10 MeV/u. A key problem in this matter are intrinsic stresses, which usually cause the thicker DLC film to curl after floating from the substrate. We solved this problem to some extent, by thermal treating the thicker films still on their slides at 770 K, which resulted in significantly reduced stresses. However, fabrication of thick stress-free DLC foils requires some more investigations.

2.2. Fabrication of large-size DLC foils

Setting up of an ERDA-ToF spectrometer for IBA with improved (due to enlarged solid angle) sensitivity at the Forschungszentrum Rossendorf required large-size and still very thin (<1 μg/cm^2) foils for the stop detector. Our relevant development efforts were directed towards producing sufficiently large DLC films, and adequate meshed frames as well. As floating and mounting of large-size DLC foils place more stringent requirements upon their mechanical strength, the film deposition rate had been reduced to $\approx 0.7\,\mathrm{ng\,cm^{-2}\,s^{-1}}$, enabling well controlled fabrication of very thin and mechanically strong foils due to reduced intrinsic stresses.

A supporting mesh is of great importance for optimal performance and rigidity of ultra-thin carbon foils. Especially, the mesh should be very flat, smooth and stretched properly over the frame. Very thin foils obviously require smaller mesh size to be mechanically stable. This however might decrease their transmission. We utilized a commercially available copper mesh with a distance of 50 μm between grid wires and a transmittance of $\approx 90\%$ which appeared to be a reasonable compromise between minimal thickness and maximal transmission of DLC target foils. However, since crosspieces are only ~ 2 μm wide in such a mesh, it is a challenge to mount a mesh over the frame properly. To overcome this difficulty, the required meshed frames were heat treated at 770 K. This might create tension in mesh due to crystallographic transformation. That is why the mesh was contracted and stretched strongly over the frame. To avoid breaking of the meshes during annealing, frames of nickel or cobalt-based alloys (Kovar, Nichrom) with their low thermal expansion coefficients are applied. As a result, exceptionally flat and rugged meshed frames can be produced, allowing the fabrication of 0.6 μg/cm^2 thin DLC foils with a large active diameter of 66 mm. The thermal treatment technique, mentioned above, is currently applied to produce all mesh supported DLC foils. A picture of the

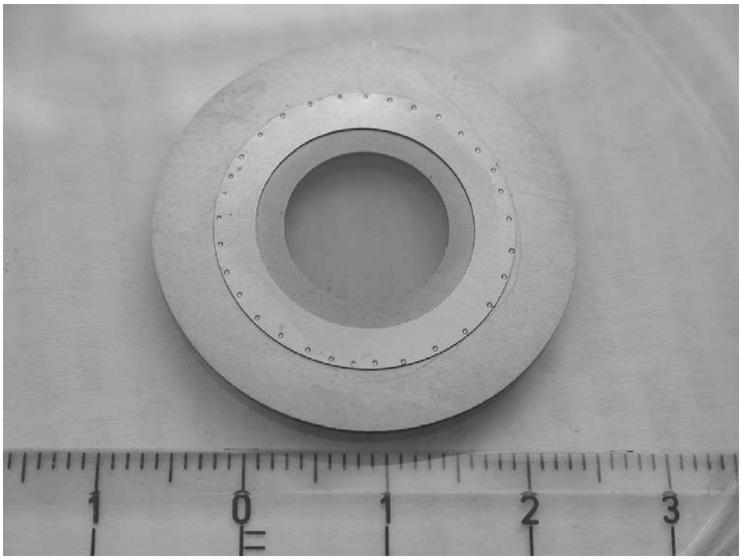

Fig. 1. Mesh-supported ultra-thin DLC start foil.

ultra-thin DLC target foil produced by this method is shown in Fig. 1.

2.3. Comparative lifetime tests of the DLC stripper foils in the several MeV energy range

Further comparative lifetime tests of thin DLC stripper foils have been carried out in moderate energy range at the Lund Pelletron. The 3 μg/cm^2 self-supporting foils were mounted on frames with a 9 mm diameter aperture and installed in the terminal together with conventional carbon foils of approximately the same thickness. No slackening of the foils was undertaken.

A 2.8 MeV $^{80}Se^{-}$ ion beam with intensities of $\leqslant 1.5$ μA and a beam spot of ≈ 5 mm diameter were used for these lifetime tests. Fig. 2 exhibits a selection of measured lifetimes (ion transmission as a function of time) obtained for beam intensities of 0.3 μA (Fig. 2a) and 1.5 μA (Fig. 2b), respectively. Fig. 2a demonstrates a nearly constant ion transmission until a first drop due to a crack in the foil caused by irradiation induced shrinkage. We observed such a stable ion transmission of DLC stripper foils with ion beams of up to 1 μA, while for higher beam intensities the ion transmission decreased gradually, as shown in Fig. 2b, perhaps due to pinhole formation caused by ion sputtering.

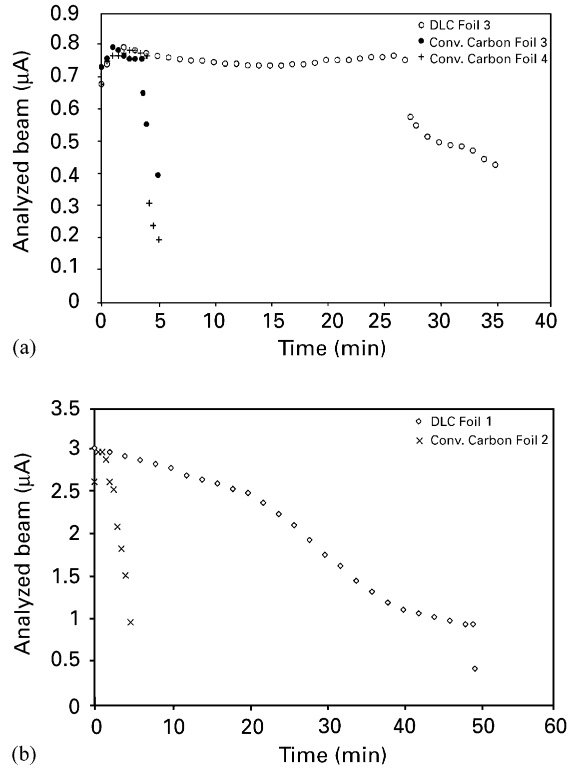

Fig. 2. Ion transmission plots obtained with DLC foils and conventional carbon stripper foils when irradiated with a 2.8 MeV $^{80}Se^{-}$ ion beam: (a) beam intensity, 0.3 μA; (b) beam intensity, 1.5 μA.

Under all irradiation conditions, DLC stripper foils lasted nearly 10 times longer than conventional carbon foils. Irradiation lifetimes as well of DLC as of conventional carbon foils were found to strongly depend on thickness in these measurements.

Approximately the same improvement in stripper foil lifetime was obtained for $10\,\mu g/cm^2$ DLC stripper foils in comparison to conventional carbon foils when exposed to 1.5 MeV $^2D^-$ beams at the tandem accelerator of the Massachusetts Institute of Technology (MIT).

2.4. Application of ultra-thin DLC foils to CEI

Ultra thin DLC foils were tested as stripping targets for molecular structure studies using the CEI method at the Heidelberg CEI setup. The multiple scattering (MS) of 5.9 MeV O^+ and 1 MeV H_2^+ ion beams penetrating DLC foils with nominal thickness of $0.6\,\mu g/cm^2$ was measured and compared to that of Formvar foils which have been used in the CEI setup so far. The detailed description of this experiment is given in Ref. [15]. Measured charge state resolved MS distributions of ions emerging in the foils have been compared to Monte-Carlo simulations. A comparison of the MS distributions of O^{5+} ions for the DLC and Formvar foils is shown in Fig. 3 together with the fitted model functions. Thus estimated effective MS thickness of 0.7–$0.8\,\mu g/cm^2$ of the DLC targets was found to be close to those of the thinnest existing Formvar foils. But DLC foils exhibit much less pin holes than Formvar foils of the same thickness and thus increase the efficiency which is especially important in experiments with molecular beams of low intensity. Other advantages of DLC foils for CEI application are smaller linear thickness due to their higher density, better uniformity and reproducibility of the production process.

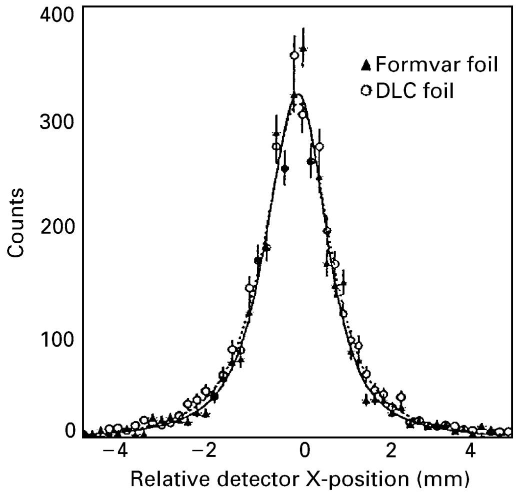

Fig. 3. Comparison of the MS distribution of 5.9 MeV $^{16}O^+$ ions for a $0.6\,\mu g/cm^2$ DLC foil and a Formvar targets of similar thickness. The fitted model functions are represented by a solid (Formvar) and a dashed (DLC) line.

2.5. Ultra-thin DLC foils for the investigation of pre-equilibrium interaction of slow ($v < v_{Bohr}$), highly charged ions with solids

The availability of ultra-thin DLC foils opens new opportunities in the investigation of fundamental processes in the interaction of slow, highly charged ions (SHCI) with solids. SHCI (<20 keV/u) such as Xe^{52+} and Au^{69+} are characterized by charge states far above of the equilibrium charge state that ions develop when travelling in solids at velocities below the Bohr velocity. Beams of SHCI are extracted from an Electron Beam Ion Trap (EBIT) and impinge on solid targets with kinetic energies ranging from 1 keV up to a few MeV [16]. Thin DLC foils allow the investigation of pre-equilibrium energy loss and charge state equilibrium dynamics [17]. The transport of "hollow atoms" from Th^{75+} ions through a foil target was recently demonstrated for the first time using a $\approx 1.4\,\mu g/cm^2$ DLC foil [18]. For these measurements DLC foils are important because their electrical resistivity is much larger than the one for conventional carbon foils and thus the reduced availability of free electrons results in increased charge state relaxation times. Hollow atoms are formed in the course of charge state relaxation of SHCI in solids, and the ability to extract them into vacuum using thin DLC foils opens a new venue for investigation of atomic relaxation processes very far from the equilibrium.

3. Conclusion

Further development and application of thin DLC target foils in accelerator experiments are planned. The comparative lifetime measurement with 3 μg/cm^2 DLC stripper foils for tandem accelerators of light and heavy ions in the MeV energy range have shown improvement factors of about 10 which is close to that for higher energy heavy-ion tandems. Fabrication of thicker (up to 200 μg/cm^2) stripper foils for higher energy linacs is within reach. Expanding of DLC targetry to improved ToF-ERDA systems for IBA analysis resulted in development of enlarged (66 mm diameter) ultra-thin SE emitting foils. A novel thermal treatment technique for the fabrication of meshed frames with improved flatness and mechanical stability made it possible to produce such large and very thin DLC foils. Ultra-thin DLC targets have shown to be advantageous for the Coulomb explosion imaging (CEI) technique. For the first time, the effective thickness of ultra-thin DLC foils was measured by means of small-angle MS and found to be close (30%) to the nominal thickness of the foil, as measured during its preparation. The availability of ultra-thin "dielectric-like" DLC targets with high electrical resistivity opens new opportunities in the investigation of pre-equilibrium ion–solid interactions.

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